

**ANALYSIS OF ASIAN OUTFLOW OVER THE WESTERN PACIFIC
USING OBSERVATIONS FROM TRACE-P**

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1. INTRODUCTION

The Transport and Chemical Evolution over the Pacific (TRACE-P) aircraft mission was conducted in February-April 2001 by the NASA Global Tropospheric Experiment (GTE) to observe the chemical outflow from Asia to the Pacific and relate it quantitatively to its sources. It used two aircraft, a DC-8 and a P-3B, based in Hong Kong and at Yokota Air Force Base (AFB) near Tokyo. It was the latest in a 20-year series of GTE aircraft missions focused on the chemistry of the global troposphere. Industrialization of the Asian continent is expected to be a major driver of future global change in atmospheric composition. Most of the outflow from the Asian continent is to the Pacific. GTE had previously conducted a series of Pacific Exploratory Missions (PEM) including PEM-West A and B (Aug-Sep 1991 and Feb-Mar 1994) over the NW Pacific, and PEM-Tropics A and B (Sep-Oct 1996 and Mar-Apr 1999) over the tropical and South Pacific. The PEM-West missions offered a first characterization of Asian outflow to the Pacific. The task of TRACE-P was to go beyond PEM-West in providing a quantitative understanding of the outflow, its origin, and its evolution.

TRACE-P was organized around two objectives. The first was "to determine the chemical composition of the Asian outflow over the western Pacific in spring in order to understand and quantify the export of chemically and radiatively important gases and aerosols, and their precursors, from the Asian continent". This objective defined a broad chemical scope for the mission encompassing long-lived greenhouse gases, aerosols and their precursors, and oxidants and their precursors. It responded to the need for an integrated multi-species approach towards addressing climate change and global air pollution issues. It also aimed to take advantage of the improved constraints on sources offered by concurrent measurements of a large number of chemical species with overlapping origins.

The second objective of TRACE-P was "to determine the chemical evolution of the Asian outflow over the western Pacific in spring and to understand the ensemble of processes that control this evolution". This objective recognized the importance of near-field chemical aging in determining global Asian influence on ozone, aerosols, and other relatively short-lived species. It responded to the need to improve our understanding of radical chemistry and aerosol dynamics in the complex Asian outflow characterized by high concentrations of aerosols, mixing of fossil fuel, biofuel, and biomass burning influences, and interweaving of polluted and stratospheric air.

Three-dimensional chemical transport models (CTMs) played a central role in guiding the design and execution of TRACE-P. This represented a major departure from previous GTE missions, whose objectives were more exploratory. In the case of TRACE-P, it was recognized that CTMs would play a critical role in addressing quantitatively the mission objectives, and an important issue in mission planning was to optimize the value of the data set for constraining and evaluating these models. Several CTMs were engaged during mission design to predict the dominant source regions and processes contributing to chemical outflow to the Pacific, and these predictions guided the selection of operational sites and flight plans. The same CTMs were then engaged during the mission to provide daily chemical forecasts, which were used together with more standard meteorological forecasts for day-to-day flight planning. Customized "bottom-up" emission inventories for East Asia in 2000, based on best available knowledge of

socioeconomic data and emission factors, were generated by David Streets (ANL) as input to the CTMs for the above exercises. These inventories were then used after the mission as *a priori* knowledge to be tested with the “top-down” constraints provided by the aircraft observations.

Another distinguishing element of TRACE-P was the extensive linkage with observations from other platforms. Joint flights were conducted with the ACE-Asia aircraft mission based out of Iwakuni (Japan) and focused on characterizing the chemical and radiative properties of Asian aerosols over the western Pacific. Joint CTM forecasts were used to support the PHOBEA-II aircraft mission based out of Seattle (U.S. west coast), which focused on the transpacific transport of ozone, CO, and aerosols and the related interannual variability through comparison with 1999 PHOBEA observations. Effort was made to integrate the TRACE-P aircraft data with satellite observations, in particular the MOPITT measurements of carbon monoxide (CO) aboard the Terra satellite, for improved characterization of the Asian outflow.

2. PROJECT OBJECTIVES

Our analysis of the TRACE-P data focused on answering the following questions:

- How do anthropogenic sources in Asia contribute to chemical outflow over the western Pacific in spring?
- How does biomass burning in southeast Asia contribute to this outflow?
- How can the TRACE-P observations be used to better quantify the sources of environmentally important gases in eastern Asia?

Our strategy drew on a combination of data analysis and global 3-D modeling, as described below. We also contributed to the planning and execution of TRACE-P through service as mission scientist and by providing chemical model forecasts in the field.

3. RESULTS

Our analysis of the TRACE-P data has produced a number of Harvard-led papers in the research literature acknowledging support from this grant. These are summarized below and the full papers are appended to this report. In addition, we have collaborated on research papers led by other TRACE-P investigators and on which we are co-authors. These include studies by J.H. Crawford and E.V. Browell at NASA/Langley, H.E. Fuelberg, at FSU, C. Cantrell at NCAR, and H.B. Singh at NASA/Ames.

3.1 The Transport and Chemical Evolution over the Pacific (TRACE-P) aircraft mission: design, execution, and first results, by Jacob, D.J., J.H. Crawford, M.M. Kleb, V.S. Connors, R.J. Bendura, J.L. Raper, G.W. Sachse, J.C. Gille, L. Emmons, and C.L. Heald, *J. Geophys. Res.*, 108, 9000, 2003.

This paper provided the mission overview. The NASA Transport and Chemical Evolution over the Pacific (TRACE-P) aircraft mission was conducted in February–April

2001 over the NW Pacific (1) to characterize the Asian chemical outflow and relate it quantitatively to its sources and (2) to determine its chemical evolution. It used two aircraft, a DC-8 and a P-3B, operating out of Hong Kong and Yokota Air Force Base (near Tokyo), with secondary sites in Hawaii, Wake Island, Guam, Okinawa, and Midway. The aircraft carried instrumentation for measurements of long-lived greenhouse gases, ozone and its precursors, aerosols and their precursors, related species, and chemical tracers. Five chemical transport models (CTMs) were used for chemical forecasting. Customized bottom-up emission inventories for East Asia were generated prior to the mission to support chemical forecasting and to serve as a priori for evaluation with the aircraft data. Validation flights were conducted for the Measurements Of Pollution In The Troposphere (MOPITT) satellite instrument and revealed little bias ($6 \pm 2\%$) in the MOPITT measurements of CO columns. A major event of transpacific Asian pollution was characterized through combined analysis of TRACE-P and MOPITT data. The TRACE-P observations showed that cold fronts sweeping across East Asia and the associated warm conveyor belts (WCBs) are the dominant pathway for Asian outflow to the Pacific in spring. The WCBs lift both anthropogenic and biomass burning (SE Asia) effluents to the free troposphere, resulting in complex chemical signatures. The TRACE-P data are in general consistent with a priori emission inventories, lending confidence in our ability to quantify Asian emissions from socioeconomic data and emission factors. However, the residential combustion source in rural China was found to be much larger than the a priori, and there were also unexplained chemical enhancements (HCN, CH₃Cl, OCS, alkyl nitrates) in Chinese urban plumes. The Asian source of CCl₄ was found to be much higher than government estimates. Measurements of HCN and CH₃CN indicated a dominant biomass burning source and ocean sink for both gases. Large fractions of sulfate and nitrate were found to be present in dust aerosols. Photochemical activity in the Asian outflow was strongly reduced by aerosol attenuation of UV radiation, with major implications for the concentrations of HOx radicals. New particle formation, apparently from ternary nucleation involving NH₃, was observed in Chinese urban plumes.

3.2 Transport pathways for Asian combustion outflow over the Pacific: Interannual and seasonal variations, by Liu, H.Y., D.J. Jacob, I. Bey, R.M. Yantosca, B.N. Duncan, and G.W. Sachse, *J. Geophys. Res.*, **108**, 8786, 2003.

The meteorological pathways contributing to Asian pollution outflow over the Pacific were examined with a global three-dimensional model analysis of CO observations from the TRACE-P aircraft mission. The model was used also to place the TRACE-P observations in an interannual (1994–2001) and seasonal context. The major process driving Asian pollution outflow in spring is frontal lifting ahead of southeastward-moving cold fronts (the leading edge of cold surges) and transport in the boundary layer behind the cold fronts. Orographic lifting over central and eastern China combines with the cold fronts to promote the transport of Chinese pollution to the free troposphere. Outflow of seasonal biomass burning in Southeast Asia during spring takes place mostly by deep convection but also by northeastward transport and frontal lifting, mixing with the anthropogenic outflow. Boundary layer outflow over the western Pacific is largely devoid of biomass burning influence. European and African (biomass burning)

plumes in Asian outflow during TRACE-P were weak (<60 ppbv and 20 ppbv CO, respectively) and were not detectable in the observations because of superposition of the much larger Asian pollution signal. Spring 2001 (La Nina) was characterized by unusually frequent cold surge events in the Asian Pacific rim and strong convection in Southeast Asia, leading to unusually strong boundary layer outflow of anthropogenic emissions and convective outflow of biomass burning emissions in the upper troposphere. The Asian outflow flux of CO to the Pacific was found to vary seasonally by a factor of 3–4 (maximum in March and minimum in summer). The March maximum results from frequent cold surge events and seasonal biomass burning emissions.

3.3 Biomass burning emission inventory with daily resolution: application to aircraft observations of Asian outflow, by Heald, C.L., D.J. Jacob, P.I. Palmer, M.J. Evans, G.W. Sachse, H.B. Singh and D.R. Blake, *J. Geophys. Res.*, 108, 8811, 2003.

We developed a daily-resolved global emission inventory for biomass burning in support of TRACE-P using AVHRR satellite observations of fire activity corrected for data gaps and scan angle biases. We implemented this inventory in a global three-dimensional model (GEOSCHEM) to simulate aircraft CO observations during the TRACE-P mission. Seasonal biomass burning in SE Asia was a major contributor to the outflow of Asian pollution observed in TRACE-P and showed large day-to-day fluctuations that vary depending on location. Three simulations were conducted with the same 3-month total (February–April) emissions but different temporal distributions: 2001 daily resolved, 2001 monthly resolved, and climatological monthly resolved. The effect of daily resolved versus monthly resolved 2001 emissions in the simulation of CO is less than 8 ppbv in Asian outflow over the NW Pacific but can exceed 100 ppbv over source regions. The relatively small effect in Asian outflow reflects spatial and temporal averaging of emissions during ageing in the continental boundary layer. Significant improvement in the simulation of TRACE-P observations (as diagnosed by the resolved variance) is found when using 2001 monthly versus climatological monthly emissions, but using 2001 daily emissions does not offer further improvement.

3.4 Eastern asian emissions of anthropogenic halocarbons deduced from aircraft concentration data, by Palmer, P. I., D. J. Jacob, L. J. Mickley, D. R. Blake, G. W. Sachse, H. E. Fuelberg, and C. M. Kiley, *J. Geophys. Res.*, 108, 4753, 2003.

The Montreal Protocol restricts production of ozone-depleting halocarbons worldwide. Enforcement of the protocol has relied mainly on annual government statistics of production and consumption of these compounds (bottom-up approach). We showed here that aircraft observations of halocarbon:CO enhancement ratios on regional to continental scales can be used to infer halocarbon emissions, providing independent verification of the bottom-up approach. We applied this top-down approach to TRACE-P observations of Asian outflow and derived emissions from eastern Asia (China, Japan, and Korea). We derived an eastern Asian carbon tetrachloride (CCl₄) source of 21.5 Gg/yr, several-fold larger than previous estimates and amounting to 30% of the global budget for this gas. Our emission estimate for CFC-11 from eastern Asia is 50% higher than inventories derived from manufacturing records. Our emission estimates for methylchloroform (CHCl₃) and CFC-12 are in agreement with existing inventories. For

halon 1211 we find only a strong local source originating from the Shanghai area. Our emission estimates for the above gases result in a 40% increase in the ozone depletion potential (ODP) of Asian emissions relative to previous estimates, corresponding to a 10% global increase in ODP.

3.5 Asian outflow and transpacific transport of carbon monoxide and ozone pollution: An integrated satellite, aircraft and model perspective, by Heald, C.L., D.J. Jacob, A.M. Fiore, L.K. Emmons, J.C. Gille, M.N. Deeter, J. Warner, D.P. Edwards, J.H. Crawford, A.J. Hamlin, G.W. Sachse, E.V. Browell, M.A. Avery, S.A. Vay, D.J. Westberg, D. R.Blake, H.B.Singh, S.T. Sandholm, R.W. Talbot, H.E. Fuelberg, J. Geophys. Res., 108 (D24), 4804, 2003.

Satellite observations of carbon monoxide (CO) from the Measurements of Pollution in the Troposphere (MOPITT) instrument were combined with measurements from TRACE-P and with a global three-dimensional chemical transport model (GEOS-CHEM) to quantify Asian pollution outflow and its trans-Pacific transport during spring 2001. Global CO column distributions in MOPITT and GEOS-CHEM are highly correlated ($R^2 = 0.87$), with no significant model bias. The largest regional bias is over Southeast Asia, where the model is 18% too high. A 60% decrease of regional biomass burning emissions in the model (to 39 Tg yr⁻¹) would correct the discrepancy; this result is consistent with TRACE-P observations. MOPITT and TRACE-P also give consistent constraints on the Chinese source of CO from fuel combustion (181 Tg CO/yr). Four major events of trans-Pacific transport of Asian pollution in spring 2001 were seen by MOPITT, in situ platforms, and GEOS-CHEM. One of them was sampled by TRACE-P (26–27 February) as a succession of pollution layers over the northeast Pacific. These layers all originated from one single event of Asian outflow that split into northern and southern plumes over the central Pacific. The northern plume (sampled at 6–8 km off California) had no ozone enhancement. The southern subsiding plume (sampled at 2–4 km west of Hawaii) contained a 8–17 ppbv ozone enhancement, driven by decomposition of peroxyacetylnitrate (PAN) to nitrogen oxides (NO_x). This result suggests that PAN decomposition in trans-Pacific pollution plumes subsiding over the United States could lead to significant enhancements of surface ozone.

3.6 Characterization of transport errors in chemical forecasts from a global tropospheric chemical transport model, by Bey, I., D.J. Jacob, H. Liu, R.M. Yantosca, and G.W. Sachse, J. Geophys. Res., submitted, 2004.

On the basis of our GEOS-CHEM CO chemical forecasts conducted in support of TRACE-P, we proposed a new methodology to characterize errors in the representation of transport processes in chemical transport models. Model simulations using both forecast and assimilated meteorology were examined. Background CO concentrations were computed as a function of latitude and altitude and subsequently subtracted from both the observed and the model datasets to focus on the ability of the model to simulate variability on a synoptic scale. Different sampling strategies (i.e., spatial displacement and smoothing) were applied along the flight tracks to search for systematic model biases. Statistical quantities such as correlation coefficient and centered root-mean-square

difference were computed between the simulated and the observed fields and were further inter-compared using Taylor diagrams. We found no systematic bias in the model for the TRACE-P region when we considered the entire dataset (i.e., from the surface to 12 km). This result indicates that the transport error in GEOS-CHEM is globally unbiased, which has important implications for using the model to conduct inverse modeling studies. Using the first-look assimilated meteorology only provides little improvement of the correlation, in comparison with the forecast meteorology. These general statements can be refined when the entire dataset is divided into different vertical domains, i.e., the lower troposphere (<2 km), the middle troposphere (2-6 km), and the upper troposphere (>6 km). The best agreement between the observations and the model is found in the lower and middle troposphere. Downward displacements in the lower troposphere provide a better fit with the observed value, which could indicate a problem in the representation of boundary layer height in the model. Significant improvement is also found for downward and southward displacements in the upper troposphere. There are several potential sources of errors in our simulation of the continental outflow in the upper troposphere which could lead to such biases, including the location and/or the strength of deep convective cells as well as that of wildfires in Southeast Asia.